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Final Report

DEVELOPMENT OF A PIEZOELECTRIC MICROBALANCE FOR CONTINUOUS MEASUREMENT OF AEROSOL MASS CONCENTRATION

By: John G. Olin Gilmore J. Sem Dale L Christenson

Prepared for:

Environmental Health Service
National Air Pollution Control Administration
5710 Wooster Pike
Cincinnati, Ohio 45227 Contract CPA 22-69-83

Attention: J. Wagman
Project Officer

May 1970

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ABSTRACT

A piezoelectric aerosol particle mass concentration monitor has been constructed and tested. This instrument detects, with high time resolution, the actual mass concentration of an aerosol cloud. The instrument uses an electrostatic precipitator to deposit aerosol particles directly onto a piezoelectric quartz crystal microbalance. The piezoelectric microbalance system consists of a quartz crystal detector, a reference crystal, two oscillator circuits, a frequency mixing circuit, and readout equipment. The electrostatic precipitator has an advantage over impaction collection devices because it collects the total mass of most aerosols on the crystal surface. It is not subject to a lower particle size cut-off within the range of most natural aerosols. A digital readout automatically indicates actual mass concentration in units of $\mu gm/m^3$.

The collection efficiency of the electrostatic precipitator is obtained as a function of corona current and flow rate. The instrument achieves 100% collection efficiency at a flow rate of 1 LPM for corona currents of approximately 3μA, or greater. Experimental results show the high sensitivity of the mass concentration instrument, e.g., typical atmosphere particle mass concentrations (100 $\mu gm/m^3$) are measured with 5% error in less than 10 seconds. Detailed studies of the experimental mass sensitivity with several different aerosols, including automobile exhaust and ambient office pollution, show excellent agreement with thin-film deposition theory for piezoelectric microbalances.

Applications of the instrument include automobile particulate emissions monitoring, industrial hygiene measurements, ambient outdoor particulate mass concentration monitoring, industrial process control and source monitoring. The complete instrument is compact, allowing hand-carrying for fast, on-site measurement of particle mass loadings.

INTRODUCTION

The measurement of the concentration of total suspended particulate matter has become an important part of air pollution monitoring systems. Normally, high-volume samplers collect the particles onto filters over a 24-hour period. The operator removes the dirty filter once a day and replaces it with a carefully-weighed, clean filter. He then lets the dirty filter stabilize for a few hours in a controlled temperature and humidity chamber before he weighs the filter. The addition in weight to the filter during the sampling period divided by the total volume of air flow through the hi-vol sampler results in a mass concentration, usually expressed in units of micrograms per cubic meter ($\mu gm/m^3$).

Although well understood and in widespread use, filter sampling methods do not lend themselves to rapid, automatic concentration measurements necessary for today's pollution monitoring networks. First, urban areas need measurements about what is happening now to various pollutant concentrations. Filter sampling systems cannot indicate today's measurement until tomorrow. Thus, pollution control agencies cannot use them as a tool to control pollution on a day-to-day basis, but only as a system for keeping records. Second, measurements with filter systems are only average concentrations over the sampling period, normally 24-hours. Such measurements tell nothing about actual minute-by-minute concentration levels. Third, filter sampling is a manual method. Unattended, automatic data readout from remote sampling stations is not possible. An operator must service every network station daily, and then must take the filters back to the laboratory to weigh them.

Ideally, an instrument for measuring the mass concentration of suspended atmospheric particles would have these characteristics:

1. Automatic, unattended, reliable operation,
2. Instantaneous measurements or an average measurement over a short time, perhaps 5 or 10 minutes.
3. Electrical readout signal capable of remote transmission and recording, reading directly in units of $\mu gm/m^3$.
4. Detection of mass concentration directly rather than by correlation of parameters such as visibility with mass concentration.

Several suspended particulate concentration measuring instruments have been developed.[1-4] However, none of them measure particle mass concentration. All depend on experimental correlation of the indicated signal with actual particle mass concentration.

This report describes results from the development of a new particle mass concentration sensing instrument. The instrument detects the actual mass concentration of the particles rather than light scattering, visibility, or soiling properties, as do other concentration devices. Thus, the instrument is a primary mass-concentration indicating standard requiring no experimental correlation coefficient to relate the measured parameter with true mass concentration.

PRINCIPLE OF OPERATION

The aerosol mass concentration monitor uses a point-to-plane electrostatic precipitator to deposit particles onto a piezoelectric microbalance mass sensor. The electrostatic precipitator is nearly independent of the piezoelectric microbalance; so, it,will be discussed below after the description of the piezoelectric microbalance.

Olin and Sem[5] recently described the use of a piezoelectric microbalance to measure the mass of small particles. Other investigators developed piezoelectric microbalance theory for use in measuring the thickness of sputtered or evaporated thin films[6] and for use in detecting gaseous adsorption on appropriately coated crystals.[7]

When a quartz crystal or other piezoelectric material is placed in an electric field, the crystal becomes mechanically stressed. Conversely, if a piezoelectric material becomes mechanically stressed, electric charges of like polarity appear on certain surfaces of the material. When a piezoelectric material is made a part of an electrical driving circuit, the piezoelectric material vibrates with a precise natural frequency. The natural frequency depends on the orientation of the cut of the crystal with respect to the axis of the raw crystal, and on the thickness and density of the crystal. For a given cut, the natural frequency of the crystal depends on the mass of material which sticks sufficiently well to its surface. The addition of mass to the surface of a given crystal causes the natural frequency to decrease in direct proportion to this added mass. This is similar to the decrease in frequency of a vibrating mechanical spring-mass system when more mass is added to the system.

For the instrument described here, the piezoelectric crystal sensor is an AT-cut quartz crystal vibrating in its fundamental mode. As described in Ref. 1, AT-cut quartz crystals vibrate in the thickness shear mode. In the fundamental thickness-shear mode, the vibrational node is a plan passing through the center of the crystal, between the flat surfaces of the plate-like crystal. No node exists

on the surface where particles are weighed. Only the portion of the crystal between the electrodes vibrates. The sensor detects any particles which stick to the electrode surface. Particles sticking to the surface of the quartz outside the electrode have a negligible effect on the vibrating portion of the crystal and are therefore not detected.

A given mass of particles sticking to the electrode of a given crystal will cause a downward shift in natural vibrational frequency, which can be calculated. The simplified relationship[5] between added mass and natural frequency for AT-cut quartz crystals is:

$$\Delta f = - \frac{2.27 f_o^2}{A} \Delta M, \qquad (1)$$

where:

Δf = change in natural frequency, Hz

f_o = natural vibrational frequency of the crystal, MHz

A = electrode area of crystal, cm^2.

ΔM = mass added to electrode area, μgm

If the frequency shift Δf is sufficiently small in comparison to the natural frequency f_o (typically Δf is less than 1% of f_o), then Δf is the linear function of the added mass ΔM, given by Eq. (1). A typical crystal sensor used in tests described below has a 5 MHz natural frequency with 0.635 cm diameter electrodes. The theoretical sensitivity $S = \Delta f / \Delta M$ of this sensor is:

$$S = 179 \ \frac{Hz}{\mu gm} \qquad (2)$$

Since a frequency shift of 0.1 Hz can easily be measured with simple frequency counters, a mass of approximately 0.005 μgm can be detected with this crystal.

In the instrument described here, a point-to-plane electrostatic precipitator deposits the airborne particles onto the piezoelectric crystal sensor. Figure 1 shows the precipitator and sensor configuration. The precipitator has several unique features:

1. Particles enter along the corona needle. This makes the deposit concentric. It also allows the particles to be directed through the most intense portion of the corona discharge, assuring high charging efficiency. The air flow and the electric field work together to carry particles from the high-efficiency charging area directly toward the sensor electrode, assuring high collection efficiency.
2. The nozzle leading into the precipitation chamber tapers toward the needle tip bringing the airborne particles into the highest intensity corona. This assures high efficiency charging with low losses on the chamber walls.
3. A constant-current high-voltage supply drives the precipitator. The constant corona current reduces the changes in charging and collection efficiency which might be caused by variation in needle sharpness, changes in point-to-plane distance, and other instabilities. As long as the corona current remains constant, the collection efficiency remains high.

Figure 2 shows a complete precipitator-microbalance system. An external vacuum source draws 1 LPM of contaminated air through the system. As airborne particles enter the intense corona discharge near the needle point, they become highly charged with negative polarity. A combination of air flow and electric field carry the particles to the electrode surface of the piezoelectric crystal.

The high adhesive force between the particles and the surface cause the particles to stick firmly to the surface. The electrical driving circuit responds to the increased mass on the crystal with a downward change in natural vibrational frequency of the crystal. An external frequency counter senses and displays the output frequency signal of the driving circuit. The mass concentration is then calculated as:

$$C = \frac{|f_2 - f_1|}{S\ t\ Q}\ , \qquad (3)$$

where:

C = aerosol mass concentration, $\mu gm/m^3$

f_2 = crystal frequency at time = t, Hz

f_1 = crystal frequency at time = 0, Hz

t = sample time, seconds

Q = Volumetric air sampling rate, m^3/sec

S = mass sensitivity of the crystal sensor, $Hz/\mu gm$

For the sensor described by Eq. (2) with an aerosol flow rate of 1 LPM,

$$C = 333\ \frac{|f_2 - f_1|}{t}, \qquad (4)$$

with units defined by Eq. (3). Based on a resolution of $\pm$ 0.1 Hz and an accuracy requirement of 5% the time resolution for the sensor given by Eq. (4), can be estimated as:

- Atmospheric Aerosol ($C = 100\ \mu gm/m^3$): $t = 6.66$ seconds.
- Automotive Particulate Emissions ($C = 10{,}000\ \mu gm/m^3$): $t = 0.0666$ seconds.

PRECIPITATOR DESIGN

The requirements of a device to collect particles from the air and deposit them onto the piezoelectric crystal are:

1. Must be nearly 100% efficient in collecting particles which contribute significantly to mass (approximately 0.01 - 20μ diameter for most naturally-occuring aerosols),
2. Must deposit particles uniformly on the piezoelectric crystal,
3. Must not interfere with the operation of the piezoelectric microbalance,
4. Must not significantly affect the mass of particles it deposits,
5. Must operate over a wide range of particle and gas-stream parameters, such as particle size, particle mass loading, air temperature, and air humidity,
6. Must be compact and geometrically compatible with the piezoelectric microbalance.

The requirement of being able to collect particles from 0.01 - 20μ diameter suggested an electrostatic precipitator. The uniform deposition requirement suggested a point-to-plane precipitator with aerosol entering the precipitating chamber symmetrically along the needle axis rather than from one side. The precipitator described here meets all other requirements.

Figure 1 shows a cross-sectional view of the precipitator. The sampling block is teflon, capable of withstanding gas temperatures of at least 400°F. Teflon is also an excellent electrical insulator and is inert to most chemicals found in

ambient air or auto exhaust. The needle is tungsten sharpened to a moderately sharp point. The hardness of tungsten facilitates retention of the sharpness of the needle point longer than nearly any other material. The needle is secured to a nylon screw with epoxy, allowing easy removal without exposing high voltage outside the block. Tight controls during machining and needle manufacture assure alignment of the needle near the center of the nozzle.

The passage bringing the particles into the precipitating chamber has a minimum of bends (one 30^{o} bend) and ledges minimizing aerosol loss by impaction. The passage diameter is optimized to reduce particle loss by Brownian diffusion, while also minimizing loss of larger particles. The length of the passage is short. The passage directs the aerosol upward, rather than downward, which prevents very large particles or large liquid drops, falling from overhead surfaces or loosened from inlet hoses, from entering the prcipitating chamber.

The size and shape of the precipitating chamber optimizes the collection efficiency of particles. The sensitivity of the piezoelectric microbalance depends inversely on crystal electrode area (see Eq. 1); so, the chamber has as small a diameter as possible, while still maintaining high collection efficiency. The depth of the chamber (point-to-plane distance) is large enough to allow time for particles to become both charged and precipitated, or collected, but small enough to maintain a stable corona current with reasonable voltages. Experimental results for collection efficiency agree well with detailed calibrations based on electrostatic charging and precipitating theory. The spacing between the crystal surface and the precipitator block is large enough to allow air flow but small enough

to force all particles to pass close to the crystal electrode area assuring high collection efficiency. Since the precipitating chamber is made of teflon, its inner surface carries a high surface charge with the same polarity as the particles, making deposition on the chamber walls negligible.

The high voltage power supply operates at constant corona current, assuring constant particle collection efficiency regardless of needle-point wear or point-to-plane spacing and varying air and particle conditions. A meter monitors the corona current, indicating any malfunction of the precipitator power-supply system.

PIEZOELECTRIC MICROBALANCE DESIGN

The requirements of a piezoelectric microbalance which detects the mass of particles deposited on its surface by an electrostatic precipitator are:

1. Must accurately sense the mass of particles with diameters from 0.01 to 10μ,
2. Must be electronically stable with one crystal electrode acting as the plane in a point-to-plane electrostatic precipitator,
3. Must be relatively insensitive to any reasonable environmental changes such as temperature, humidity, or gaseous composition,
4. Must be relatively insensitive to location of particle deposit on the electrode surface.
5. Must be capable of accurately sensing the mass of particles deposited on the crystal electrode while sampling mass concentrations from 10 - 100,000 $\mu gm/m^3$,
6. Must be completely compatible with a point-to-plane electrostatic precipitator,
7. Must be rugged.

The requirement of insensitivity to deposition location led to the choice of a type AT quartz crystal which has no nodes on either electrode surface. Also, quartz crystals are most common and are more stable than most piezoelectric materials. The sensitivity requirement pointed to high frequency AT crystals, but the highest frequency AT crystals are very thin and hence fragile; so, 5 MHz crystals offer a reasonable compromise.

A reference crystal corrects the piezoelectric microbalance for the effects of environmental changes. AT quartz crystals can be cut to have a very low temperature coefficient over a temperature range of about 30°C. Temperature corrections for a wider range must be made by use of a reference crystal.

The reference crystal is identical to the primary mass-sensing crystal. Except for particles, it is exposed to the same ambient conditions as the primary crystal. Physically, the reference crystal is downstream from the precipitator and primary crystal, as shown in Fig. 2. Thus, the reference crystal experiences almost the same ambient conditions as the primary crystal.

Figure 2 shows how the crystal sensors mount within the sampling head of the aerosol mass concentration monitor. The crystal holders can be pulled out of the sampling block. Small tabs, firmly attached to the crystal holder, clamp the crystal to the crystal holder and make connection to the two electrodes of the crystal. The operator can pull the crystal out of the tabs for cleaning or replacement. (Cleaning the crystal requires just a wipe with a damp cloth.) Thus, crystal cleaning, the primary maintenance requirement of the instrument, takes only a couple of minutes.

Figure 3 shows the electronic block diagram for both the primary and reference crystals. Separate electronic circuits drive the primary and the reference crystals. The frequency signals from the two driving circuits enter a frequency mixer which electrically subtracts the two frequency signals resulting in a "beat" frequency. The beat frequency is the output frequency signal of the piezoelectric microbalance. The mixer subtracts out the effect of extraneous changes in the ambient environment, such as gas temperature, adsorption of water vapor or other gaseous constituents, etc.

DIRECT CONCENTRATION READOUT

A simple, 5-digit, 0 - 100 KHz, frequency counter can measure the frequency of the mixer output signal of the piezoelectric microbalance. The frequency change during a given period of time converts directly into aerosol mass concentration by means of Eq. (4). However, this method requires the operator to make the concentration calculation after he gets the data from the frequency counter. To avoid this, an automatic readout unit was successfully designed, built and tested. It directly computes the particle mass concentration in units of $\mu gm/m^3$, thus , eliminating the need for human calculation.

Equation 4 shows that the particle mass concentration is a constant multiplied by the ratio of crystal frequency change to the sampling time. The operation of the particle mass-concentration computer is based on Eq. (4). The sampling period t is automatically preset to cancel out the constant in Eq. (4). Just before the sampling period, the computer automatically counts the frequency f_1 downward. Just after the sampling period, it counts the frequency f_2 upward starting at the value f_1. The

resulting frequency difference is equal to the particle mass concentration C. The sampling period can be preset to accommodate particle mass concentrations in any one of the following three ranges: 0 - 10^3, 0 - 10^4, and 0 - 10^5 $\mu gm/m^3$. The unit provides a digital display of either the output frequency (Hz) or the particle mass concentration ($\mu gm/m^3$). The output is digital and is compatible with standard digital printers. The unit automatically controls the sampling sequence of the electrostatic precipitator. The entire system allows measurement of particle mass concentration with a digital data readout in $\mu gm/m^3$ updated every few seconds.

EXPERIMENTAL PERFORMANCE

In evaluating the performance of the aerosol mass-concentration monitor, two basic questions require experimental answers:

1. How well does the precipitator collect the particles and deposit them onto the crystal electrode?
2. How well does the piezoelectric microbalance sense the mass of these deposited particles?

Tests to answer either question must cover the variety of conditions under which one might expect the instrument to operate. The discussion below covers the results of a number of tests which partially answer the two questions.

Precipitator efficiency depends on a number of factors. Geometrical factors include point-to-plane distance, precipitating chamber size and shape, and the size and shape of the chamber entrance and exit nozzle. Aerosol factors include particle size, shape, and composition; flow rate; mass concentration; and gas composition and pressure. Other important factors include the electrical properties of the

chamber walls and point-to-plane corona current intensity. The optimum precipitator would have a geometry and operating conditions such that it would collect any aerosol of interest on the crystal electrode with 100% efficiency.

Figure 4 shows the results of a series of efficiency experiments varying the aerosol flow rate and the corona current. The geometry of the precipitating chamber is slightly different from the final design shown in Fig. 2. Later tests with the final design using several identical operating conditions showed somewhat higher efficiences than those in Fig. 4. The aerosol was high-concentration tobacco smoke (10,000 - 100,000 $\mu gm/m^3$). Notice that with increased aerosol flow rate, 100% efficiency requires higher corona current.

This series of tests indicates that the precipitator operates with essentially 100% efficiency on a relatively high concentration of tobacco smoke. The mass of tobacco smoke is probably contained by particles in the 0.01 - 3.0μ diameter size range. Two tests with office aerosol resulted in precipitator efficiencies of 96 and 102%. Tests with several other aerosols within this size range, including auto exhaust and ambient laboratory air having lower concentrations than tobacco smoke, also showed near 100% precipitator efficiency.

For the piezoelectric microbalance to detect the particles, they must land on the crystal electrode. Examination of the crystal after numerous tests verifies that almost all particles deposited on the crystal are on the electrode. Examination of the walls of the sampling chamber, the entrance duct, and the entrance nozzle after many tests shows that very few particles deposit on surfaces other than the crystal electrode.

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When sampling certain types of aerosols, the precipitator appears to cause certain gaseous components of the air to precipitate out as particles. Apparently, the highly reactive corona discharge causes a chemical reaction resulting in a precipitate. This precipitate has been carefully analyzed and is expected to be a complex hydrocarbon formed by the polymerization in the high electric field of the corona discharge of gaseous hydrocarbon, such as oil vapors. In a series of tests in an industrial manufacturing facility, this effect appeared sporadically, ranging from 0 - 20 $\mu gm/m^3$ equivalent concentration. The test to detect this effect is to filter the inlet air with an absolute filter and operate the instrument. The produced deposit appears both as a change in frequency of the piezoelectric microbalance and as a deposit of particles visible with an optical microscope. Thus, simple corrections can be made for this effect, either by operating two mass monitors with a filter on one, and subtracting the amount on the filtered one from the other, or by measuring the effect before and after a measurement of aerosol mass concentration and assuming the effect is constant during the run. It should be emphasized that this "interference" effect is either not present, or is neglibile, (since its magnitude is less than $\pm$ 10 $\mu gm/m^3$) during sampling of most high-concentration aerosols around 1000 $\mu gm/m^3$ such as auto exhaust. The effect is only a problem occasionally when sampling certain aerosols with low concentrations. Fortunately in this case ambient gaseous hydrocarbons are also probably very low in concentration. For example, the "interference" effect was found to be negligible when sampling atmospheric air in a suburban area. Further work is needed to define this problem completely.

The ability of the piezoelectric microbalance to detect the mass of particles on its surface depends strongly on how well the particles stick to the crystal electrode surface. Particles must vibrate as if they are an integral part of the crystal before the crystal will detect them. Olin and Sem[5] described a number of experiments verifying the ability of the basic piezoelectric microbalance to detect small particles. The results below extend these experiments to the sampling of commonly-occurring aerosols with the complete aerosol mass-concentration monitoring system.

The easiest way to find whether particles are being detected by the piezoelectric microbalance is to check the experimental against the theoretical mass sensitivity of the crystal. Equations 1 and 2 define theoretical mass sensitivity S. A highly sensitive standard gravitational balance must be used to measure the mass of the deposit on a crystal.

Table 1 compares experimental and theoretical mass sensitivity from a number of experiments using a variety of crystals and aerosols. Note that all the experimental mass sensitivities agree with theoretical mass sensitivities within experimental error. Difficulty in defining the exact area of the crystal electrode causes the uncertainty in theoretical mass sensitivity. Errors in the balance used to weigh filters and crystals causes the relatively large uncertainty in experimental mass sensitivity. The balance used to obtain this experimental data is accurate to about $\pm$ 30 µgms. The large number of tests with concentrated tobacco smoke and automobile exhaust indicates that experimental mass sensitivity agrees well with theoretical mass sensitivity.

Although most tests with atmospheric and indoor aerosol indicate results well within experimental error, the statistics of a larger number of tests indicate low mass sensitivity when the relative humidity is below 30%. Apparently, water vapor adsorbed by the particles and the electrode surface significantly increases the adhesive force bonding the particles to the crystal electrode. When low humidity reduces adsorbed water, particles do not stick well enough to be sensed accurately. Preliminary tests show that passing a relatively dry aerosol through a humidifying chamber so that its relative humidity reaches 40 - 50% causes the microbalance to sense the true mass of particles. A simple inlet humidifying attachement appears to be an easy way to correct the lack of mass sensitivity for the infrequent applications in very low humidity atmospheres. For ultimate accuracy in measuring true mass concentration with such an attachment, a correction, probably a percentage of reading, may be required to account for moisture introduced artifically.

In the past, many particle mass concentration measurements have been made at convenient, standardized humidity levels. For example, in ambient air, high-volume filter sampling the filter is stablized at low humidity to correct for water vapor adsorbed on the filter material. Bringing the aerosol to an intermediate humidity of about 50% may be a more realistic measure of particle mass concentration than drying the sample out. Thus, the possible humidity limitation of the instrument does not appear to be serious, and may in fact lead to more realistic measurements of mass concentration.

The largest size of particles which stick to the electrode surface varies with the type of aerosol and the humidity of the air. For example, an experiment with dry, spherical, 10μ diameter, sand particles in air at 25% relative humidity showed poor sensitivity. On the other hand, naturally-occurring or pollutant aerosols exist in

wide size ranges and usually have water adsorbed on the surface of the particles. Often, a larger portion of the particles are partially or entirely liquid. The combination of many small particles and some liquid particles tends to augment the adhesion of larger particles in the particulate deposition to the electrode surface. Although not completely verified by tests, the largest particles detected accurately by the present piezoelectric microbalance are probably near 10μ diameter. Reference 5 discusses the adhesion of particles to the electrode surface in greater detail.

Figure 5 describes the sampling system used for a series of experiments with automobile emissions. Automobile exhaust (1.6 scfm) from an idling auto was diluted 6:1 with ambient outdoor air (9.9 scfm). A small sample of the diluted exhaust was drawn through the particle mass concentration monitor and another through a Nuclepore* filter.

Figures 6 to 9 show data resulting from two automobile exhaust experiments, one with a 1.5 MHz crystal sensor and one with a 5.0 MHz crystal. Figures 6 and 8 show the frequency output of the piezoelectric microbalance and the particle mass concentration of diluted exhaust as functions of elapsed sampling time for 1.5 and 5.0 MHz crystals, respectively. Notice the stable mass concentration measurements in Figure 6 indicated by the linear slope of the frequency line and the horizontal concentration line. The slight upward curvature of the frequency line in Figure 8 indicates a slowly increasing mass concentration. This is verified by the concentration line. The frequency output of the piezoelectric microbalance was recorded once every minute. The concentration measurement fluctuates very little from minute to minute. On other automobile exhaust experiments, data accurate to ±0.1 Hz was recorded every ten seconds. Only very small fluctuations in mass-concentration were noticeable, even for 10-second intervals. The data appeared to be just as

*Trademark of General Electric Co.

accurate and stable with 10-second recording intervals as with 1 minute intervals. A corollary conclusion to these experiments is that an automobile engine after reaching equilibrium is an excellent constant mass-concentration aerosol generator.

Both 1.5 and 5.0 MHz crystals stopped oscillating when the accumulated particle deposit reached about 400 μgm. With the 1.5 MHz crystal, 400 μgm is equivalent to about 6,000 Hz frequency shift. With the 5 MHz crystal, 400 μgm is equivalent to about 60,000 Hz frequency shift. With automobile exhaust and tobacco smoke, the piezoelectric microbalance consistently weighs over 300 μgm accumulated mass before requiring cleaning. Without inlet humidification, outdoor atmospheric aerosols have a maximum allowable crystal deposit of approximately 50 to 100 μgms.

Figures 7 and 9 compare experimental with theoretical mass sensitivity for three sets of data points taken during each of the experiments shown in Figures 6 and 8. The line in each figure represents theoretical mass sensitivity. The data points represent actual measured experimental sensitivity. Three of the data points in each figure are the result of weighing the particle deposit on the crystal at various times during the experiment. In addition, the weight of the particles on the parallel filter allows an extra mass sensitivity check point corresponding to the first crystal weighing in each figure. Additional data on the sensitivity of the filter weight is unavailable because the Nuclepore filters became overloaded. The error bars indicate possible experimental error for each data point. All of the experimental mass sensitivity data points are within experimental error of the theoretical mass sensitivity.

APPLICATIONS

The data in Figures 6 - 9 demonstrate the usefulness of the instrument for rapid, accurate measurement of the mass concentration of automobile particulate emissions. With dilution, accurate measurements of mass concentrations can be made in about 5 to 10 seconds, facilitating measurement of true mass concentration during short acceleration and deceleration portions of standard driving cycles. The unusually wide range of concentrations which the instrument can measure covers the complete range of automotive particulate emissions. The predominantly small particles present in automobile exhaust are ideal for measurement by this technique.

Another application is industrial hygiene particulate monitoring. The instrument is small and suitable as a portable pollution source monitor. Accurate particle mass-concentration measurements in most industrial or office atmospheres require just 1 to 5 minutes. Since most people spend about 90% of their time inside buildings, indoor pollution is becoming recognized as a prime pollution problem. This instrument is a tool which can help lead to control of indoor particulate pollution.

A third major application is outdoor ambient air monitoring. Pollution control officials need fast, accurate, measurements of all air pollutants to head-off major pollution episodes and to control the general pollution level. Present particulate sampling methods using filters do not let the control official know the particle mass concentration until the next day. This instrument opens the possibility of ambient particle mass concentration monitoring with accurate readout every 5 or 10 minutes. It can also be used as a portable, hand-carried, point concentration

measuring instrument. Another possible use is to measure particle mass concentration around the perimeter of a pollution source.

A fourth area of application is industrial process control. Many processes require accurate control of particle mass concentration to optimize the process. The instrument may find uses here. It also has possible application to continuous monitoring of the respirable range of particulate emissions from air-pollution sources.

CONCLUSION

A piezoelectric/electrostatic aerosol mass concentration monitor has been designed, constructed, and tested. The instrument measures particle mass,itself, not some parameter of mass. Its time resolution is high: the mass concentration of typical atmospheric aerosols can be measured with 5% accuracy in about 10 seconds and typical automotive particulate emissions in about 0.1 second. The electrostatic precipitator has 100% collection efficiency for the dense (10^5 $\mu gm/m^3$) cigarette-smoke aerosol used in the detailed collection-efficiency tests and for all other aerosols tested, including automobile exhaust, laboratory aerosols, and office aerosol. The mass sensitivity of the piezoelectric microbalance is within experimental error of its theoretical value (Eg. (1)) for the following aerosols tested: automobile exhaust, cigarette smoke, laboratory aerosol, and office aerosol.

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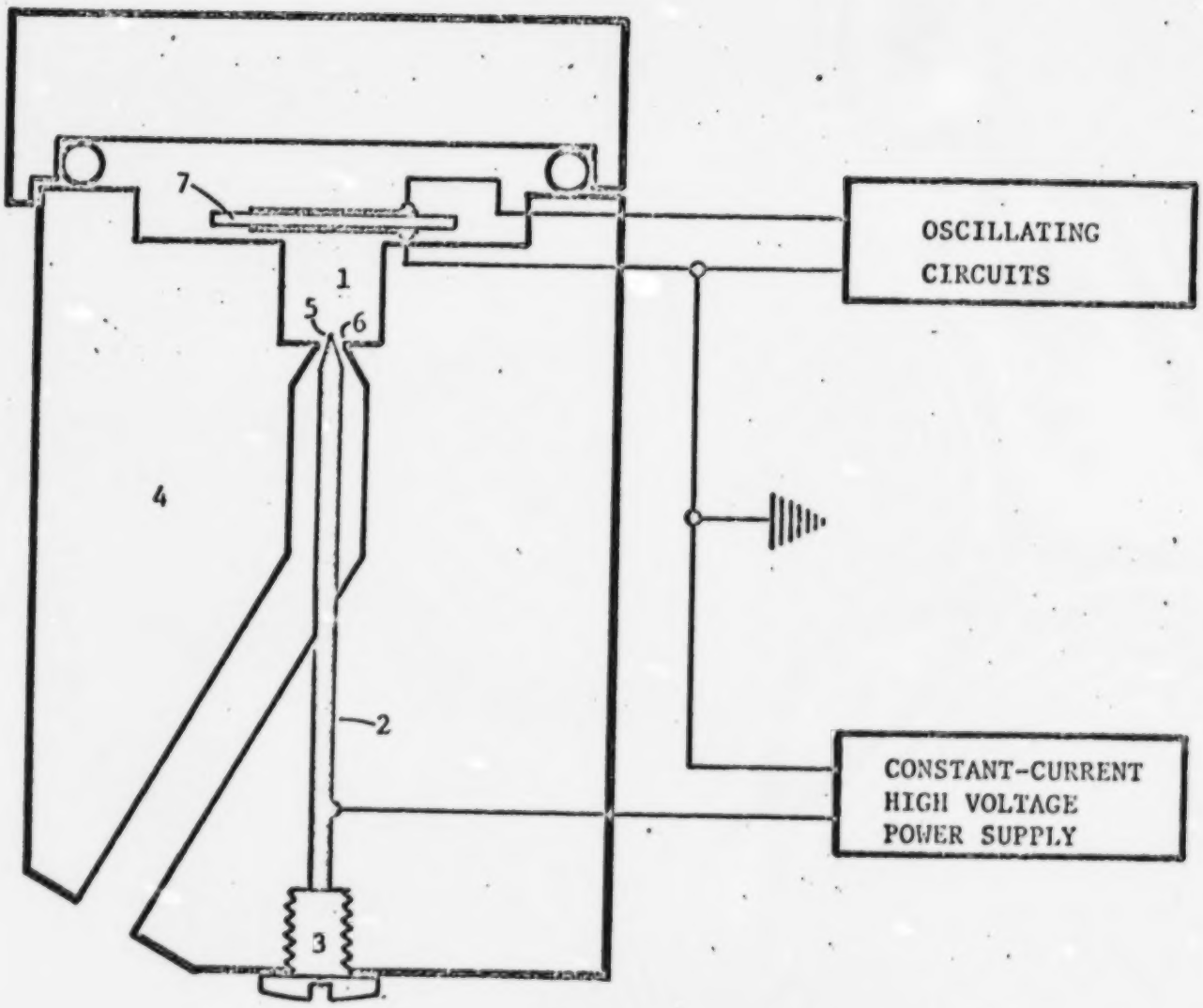

Fig. 1: Side view of electrostatic precipitator and piezoelectric microbalance: 1) precipitating chamber,1/4 in dia x 1/4 in high, 2) corona needle, 3) needle adjusting screw, 4) teflon precipitating block, 5) high corona region 6) entrance nozzle to precipitating chamber, 7) piezoelectric crystal sensor.

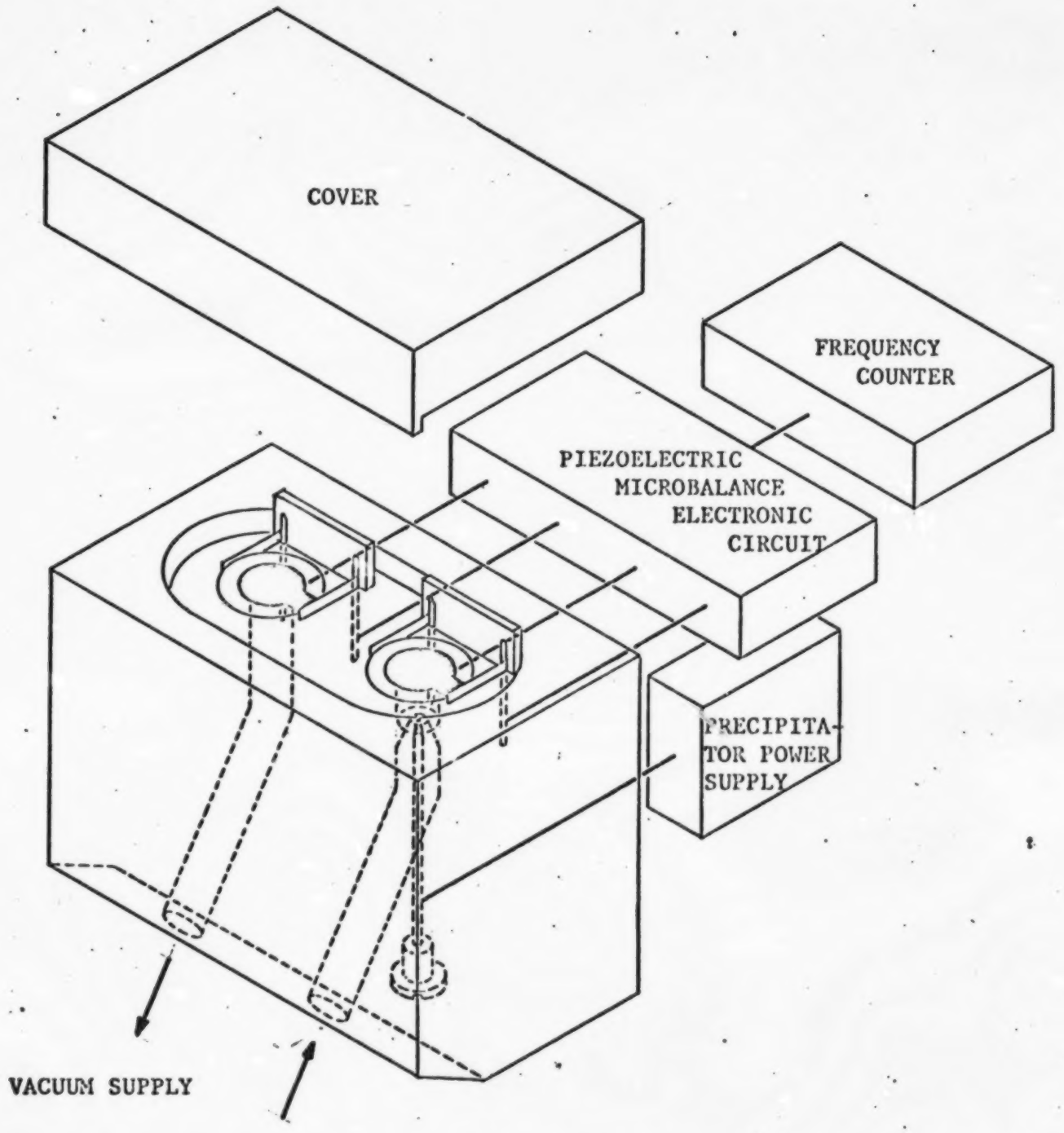

FIG. 2: Complete particle mass concentration measuring system

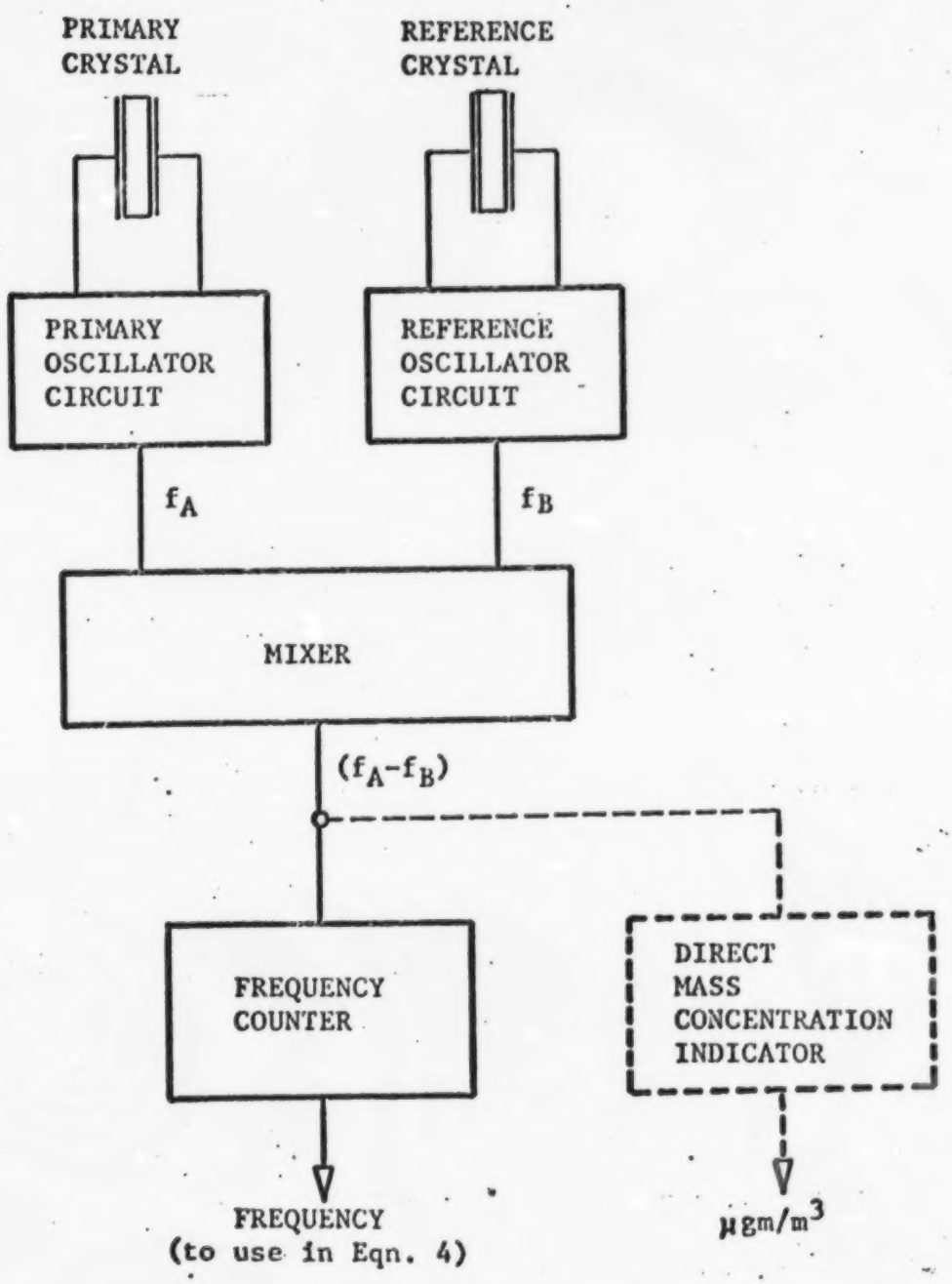

FIG. 3: Piezoelectric microbalance electronic block diagram.

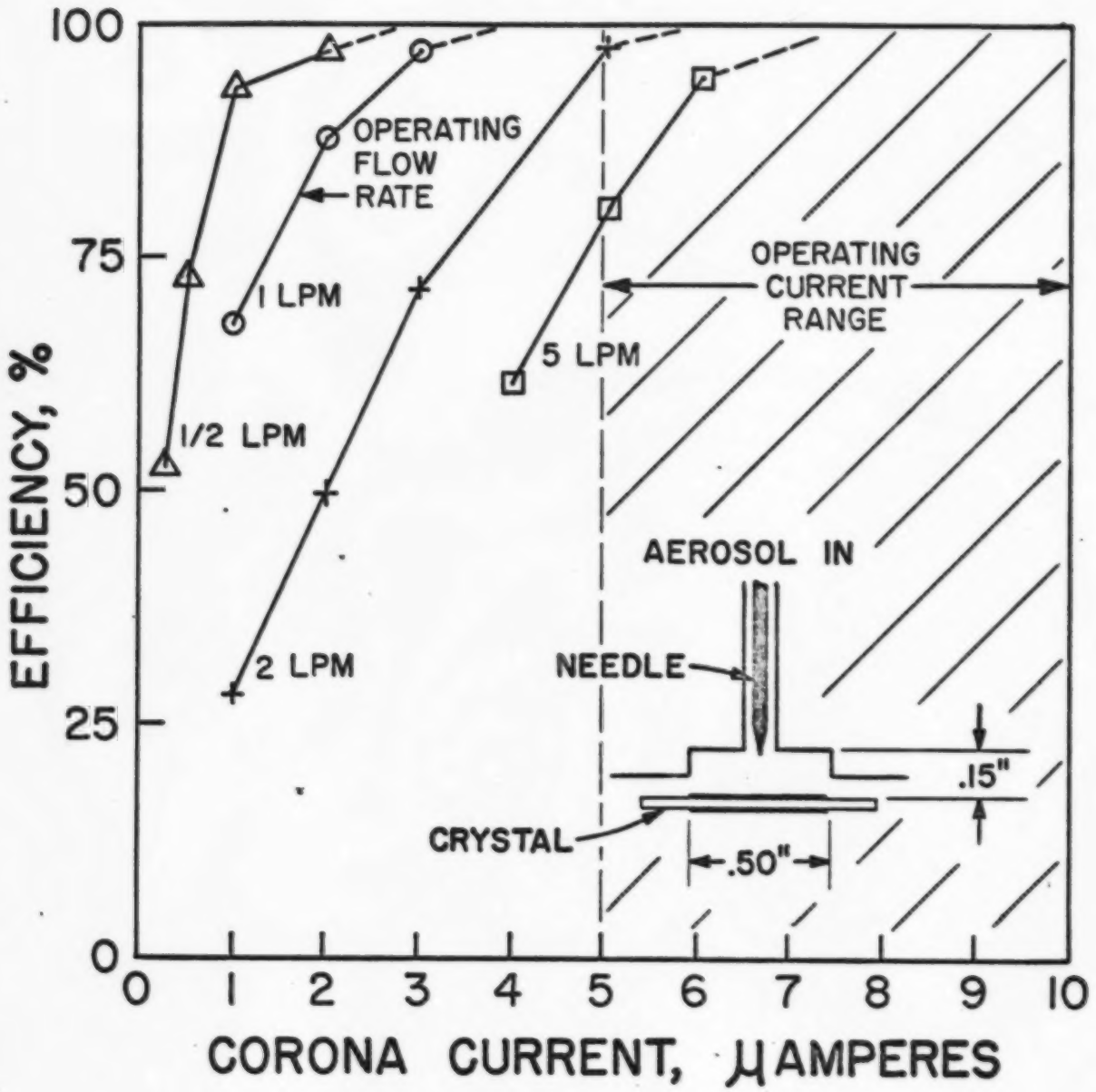

FIG. 4: RESULTS OF ELECTROSTATIC PRECIPITATOR EFFICIENCY TESTS.

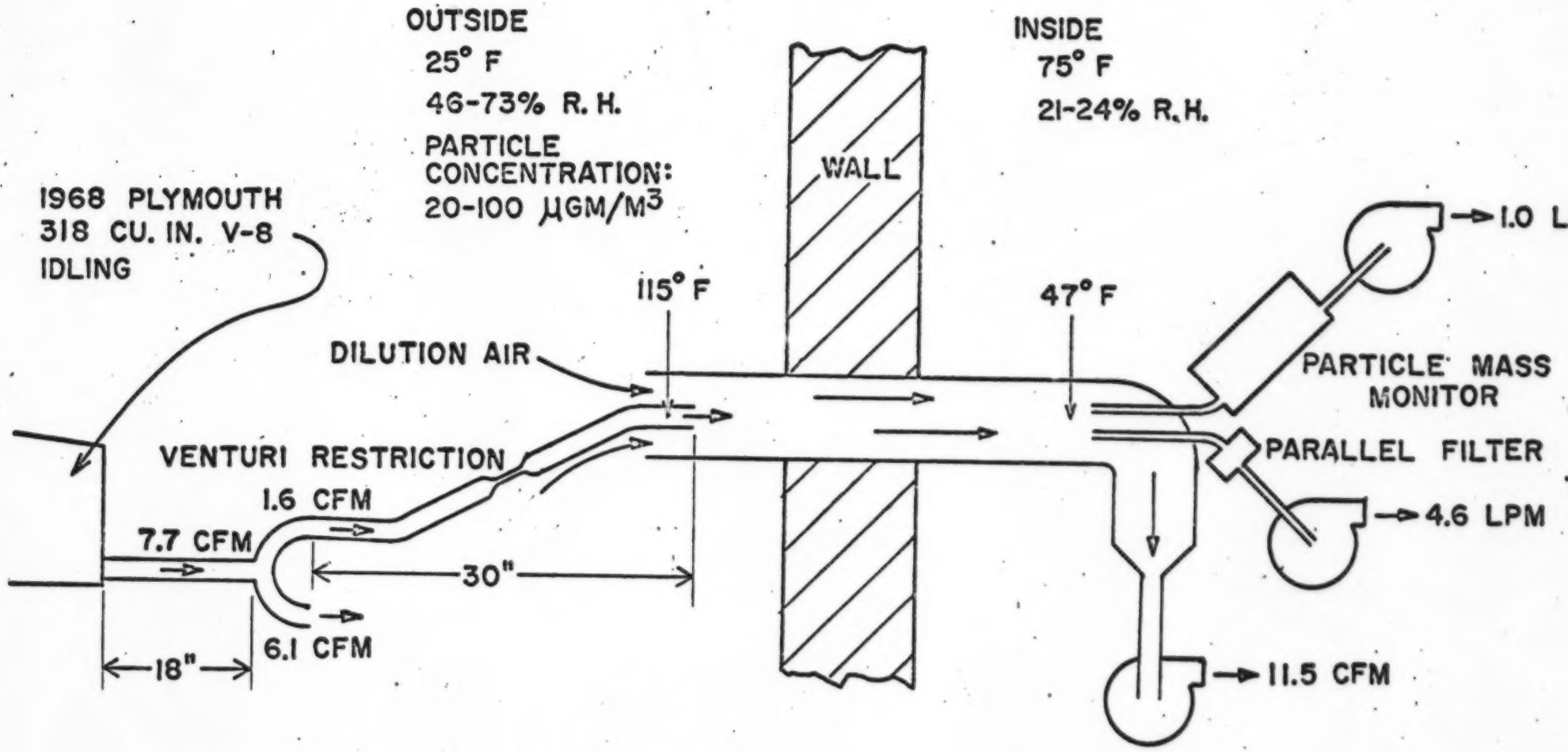

FIG. 5: Sampling system for automobile exhaust particle mass concentration tests.

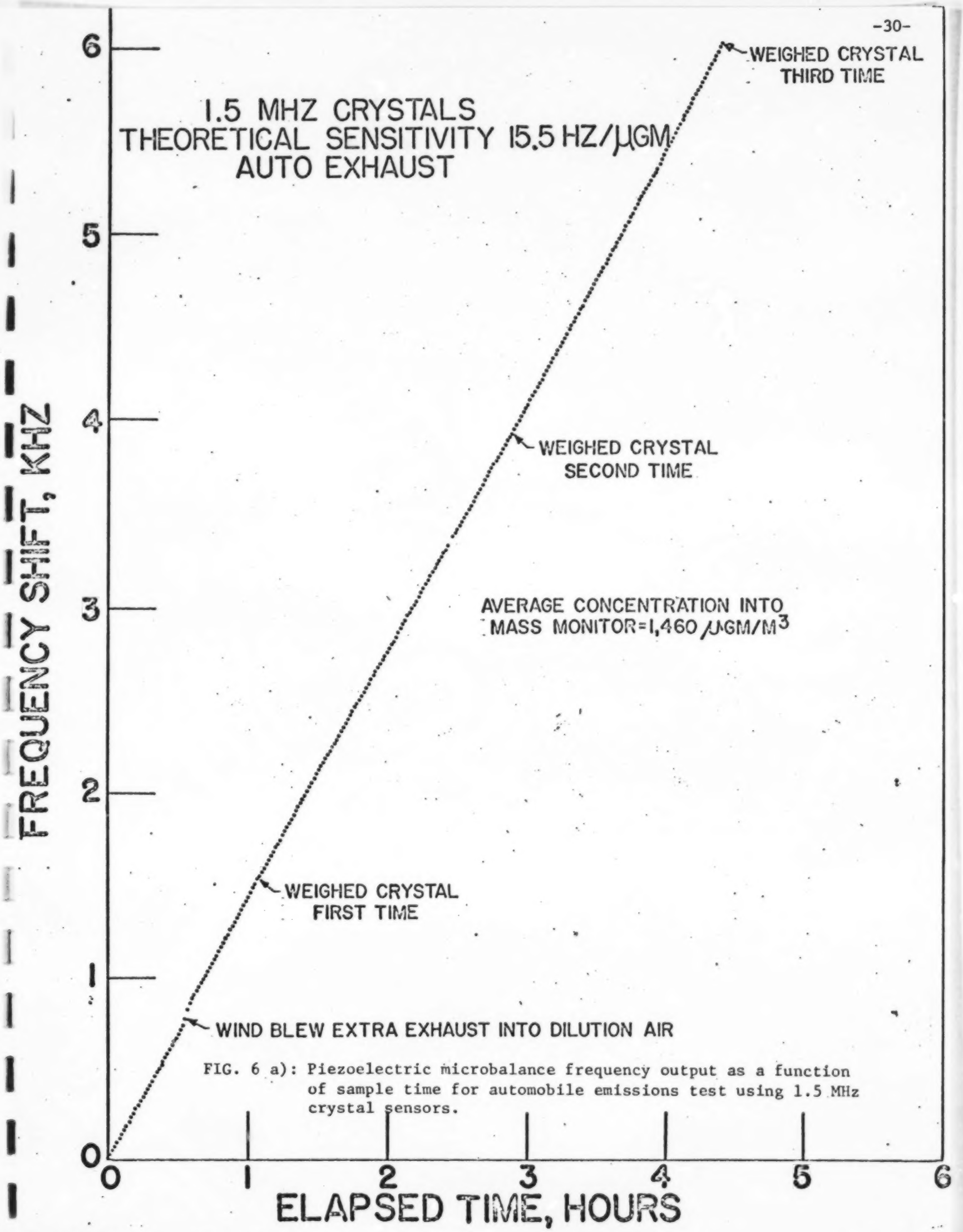

FIG. 6 a): Piezoelectric microbalance frequency output as a function of sample time for automobile emissions test using 1.5 MHz crystal sensors.

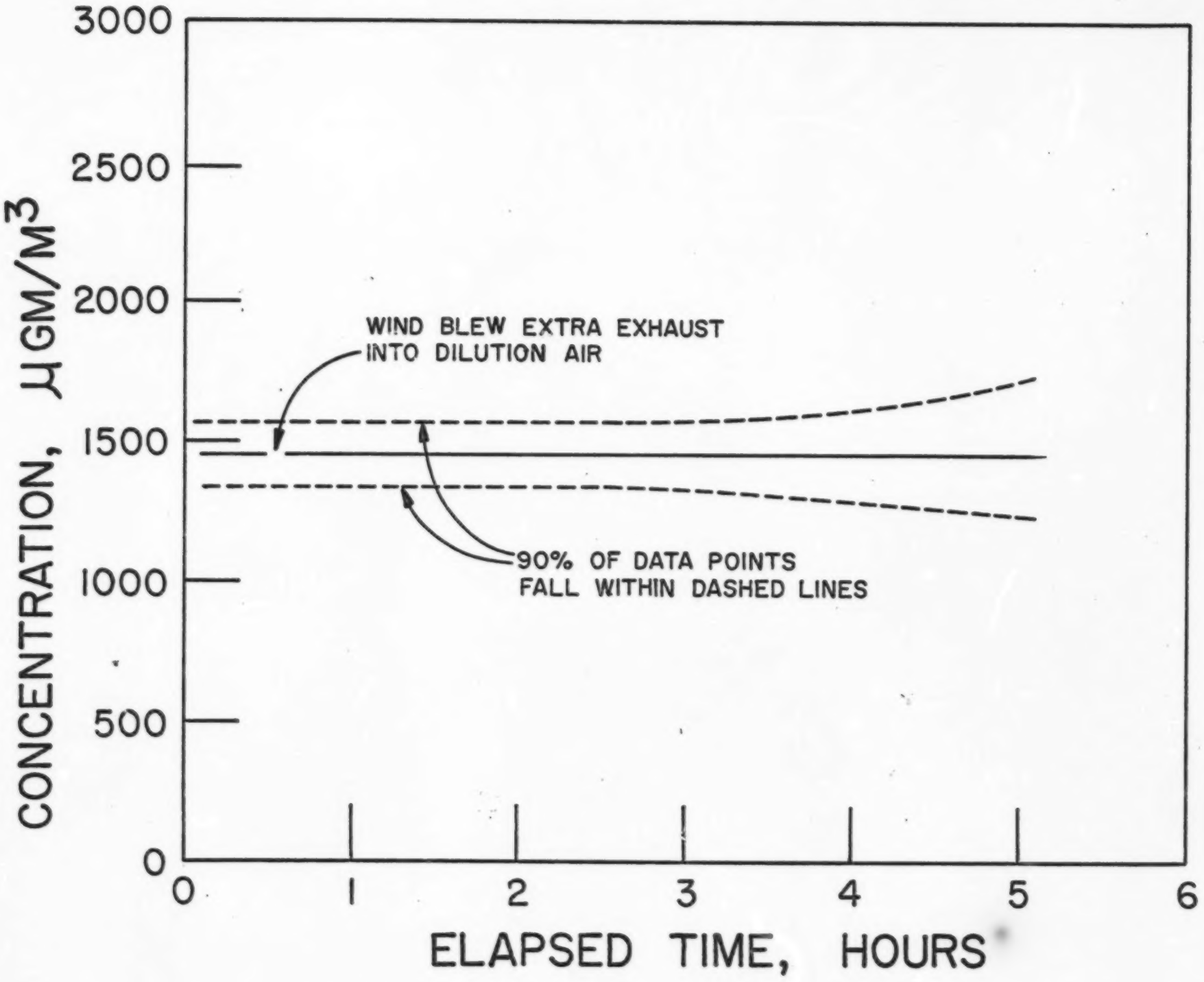

FIG. 6 b): Particle Mass Concentration as a function of sample time for automobile emissions test using 1.5 MHz crystal sensors.

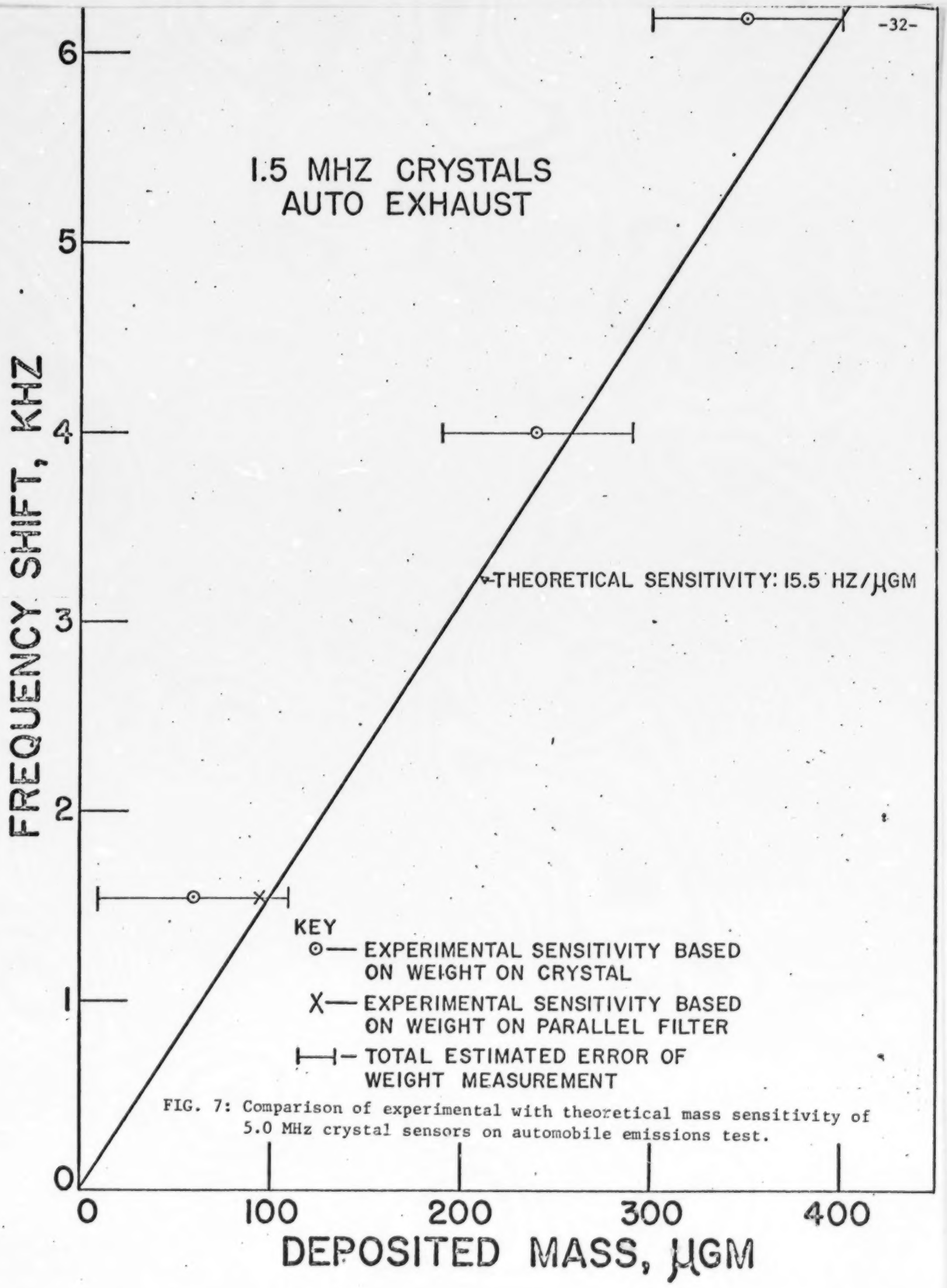

FIG. 7: Comparison of experimental with theoretical mass sensitivity of 5.0 MHz crystal sensors on automobile emissions test.

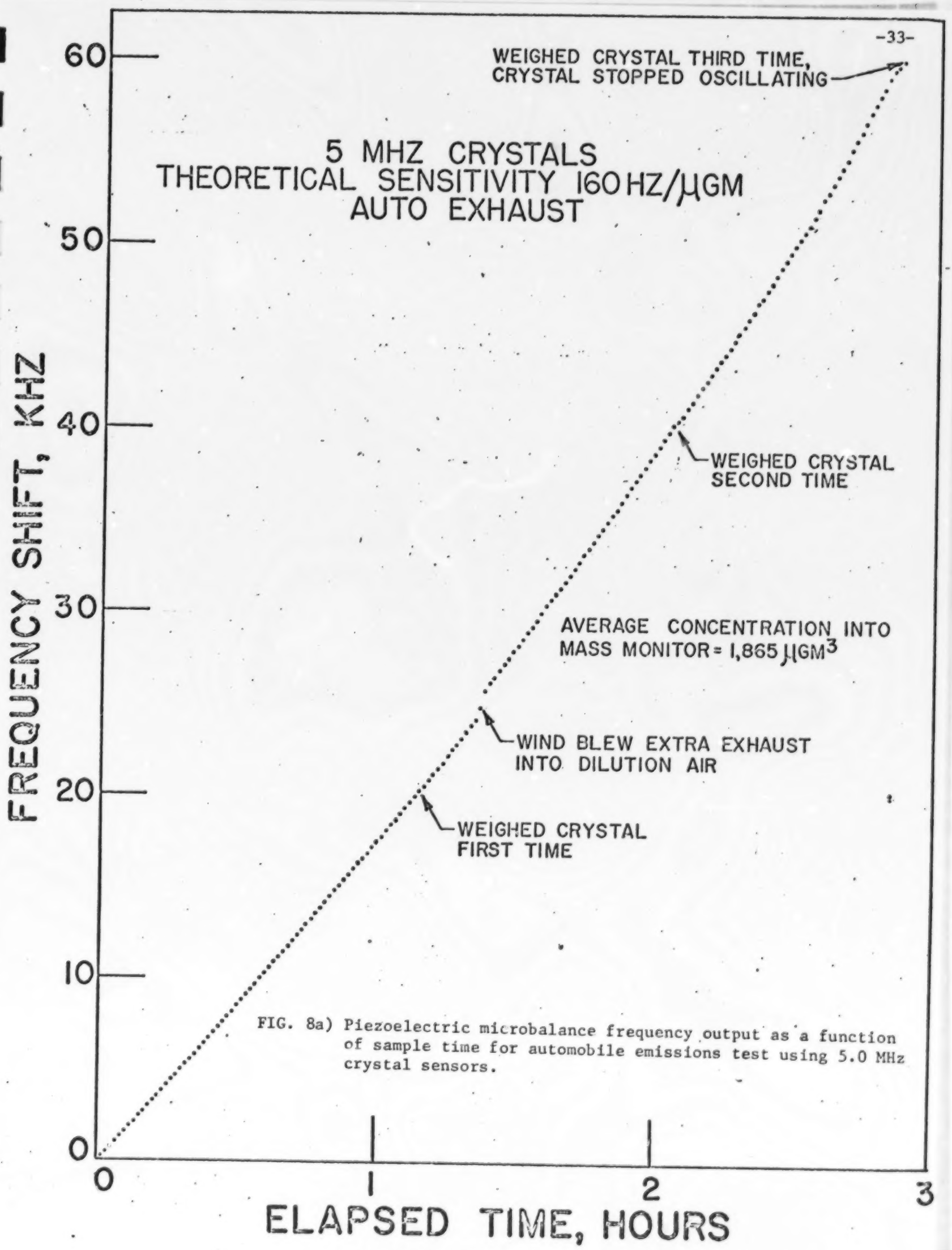

FIG. 8a) Piezoelectric microbalance frequency output as a function of sample time for automobile emissions test using 5.0 MHz crystal sensors.

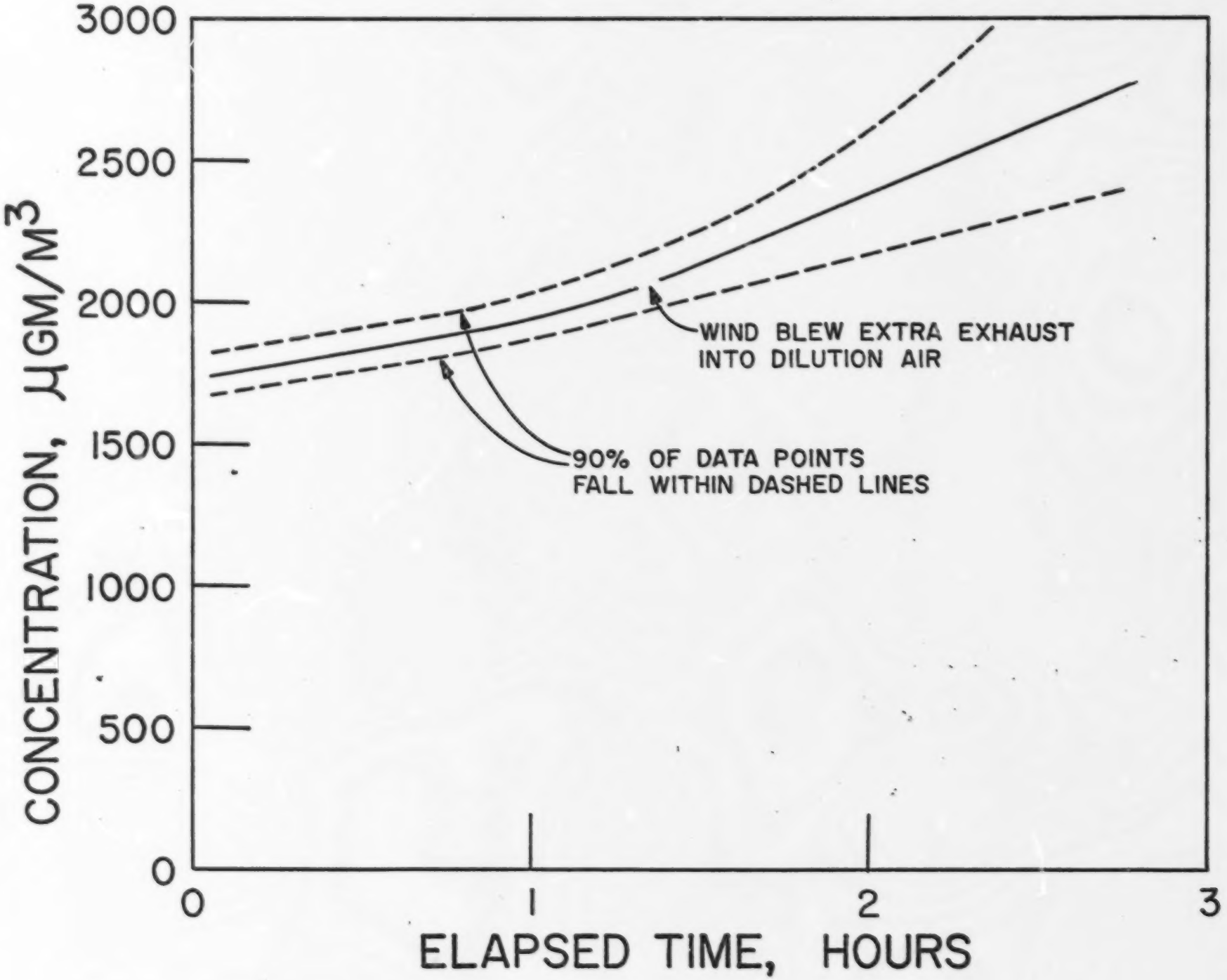

FIG. 8 b): Particle Mass Concentration as a function of sample time for automobile emissions test using 5.0 MHz crystal sensors.

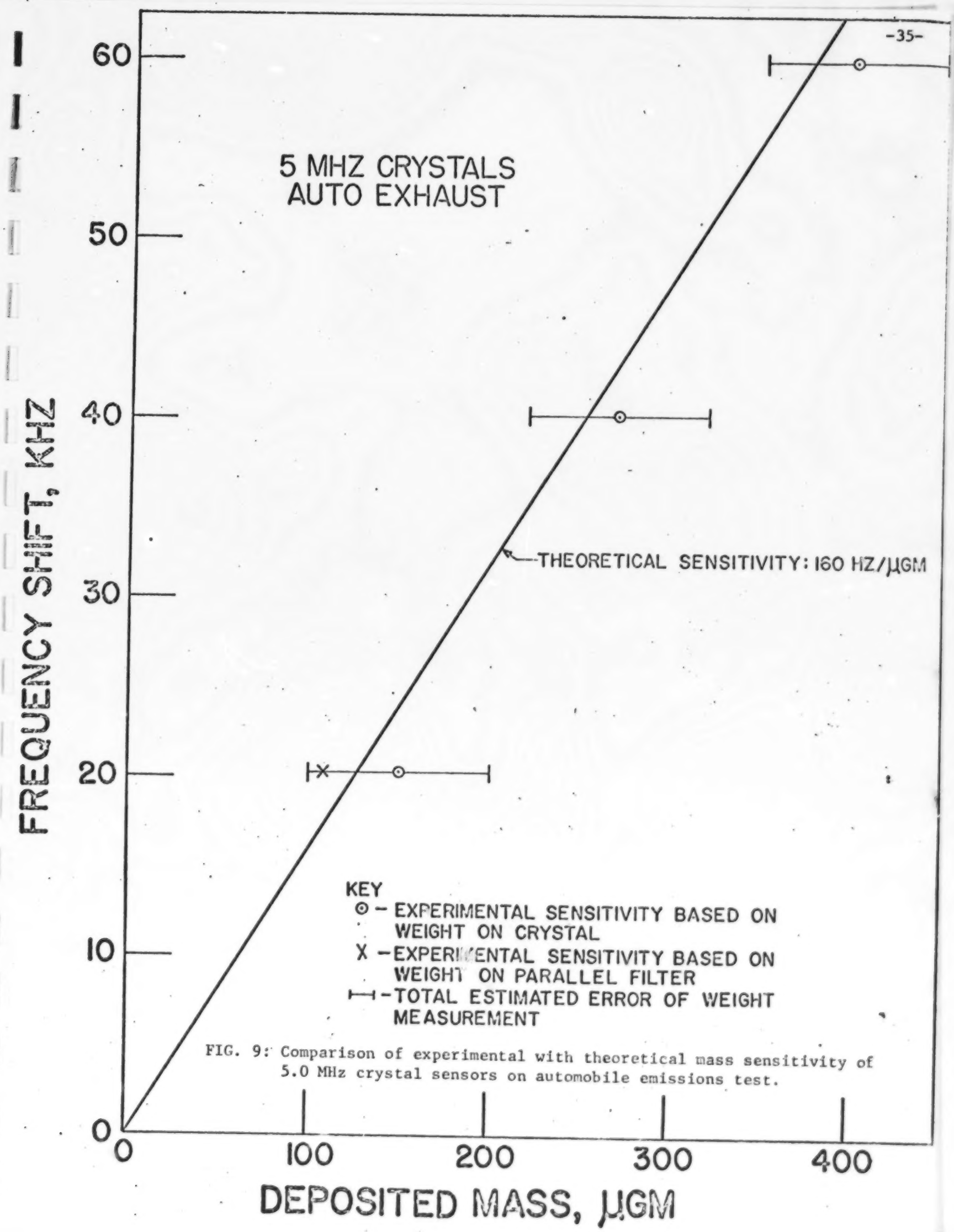

FIG. 9: Comparison of experimental with theoretical mass sensitivity of 5.0 MHz crystal sensors on automobile emissions test.

TABLE 1: Experimental mass sensitivity compared with theoretical mass sensitivity of crystals used to measure the mass concentration of several aerosols.

Crystal Frequency MHZ	Aerosol	Average Mass Conc., $\mu gm/m^3$	Mass Sensitivity, Hz/µgm Theoretical	Experimental
1.5	Tobacco smoke	14,000	15.5 ± 1	14.7 ± 2
1.5	Tobacco smoke	5,830	15.5 ± 1	14.4 ± 2
1.5	Tobacco smoke	7,230	15.5 ± 1	14.2 ± 2
5.0	Tobacco smoke	-	179 ± 10	179 ± 30
1.5	Auto exhaust	1,460	15.5 ± 1	17.6 ± 3
5.0	Auto exhaust	1,865	160 ± 10	150 ± 17
5.0	Office aerosol	133	177 ± 10	187 ± 30
5.0	Office aerosol	162	177 ± 10	198 ± 30
5.0	Office aerosol	165	177 ± 10	193 ± 30
5.0	Acrylic spray*	-	165 ± 10	158 ± 20
3.0	Acrylic spray*	-	66 ± 4	62 ± 11
1.5	Acrylic spray*	-	15.0 ± 1	16 ± 3
1.5	Ambient Laboratory Pollution**	75 - 100	15.5 ± 1	10.5 ± 6

*Acrylic was sprayed directly onto the crystal electrode from an aerosol spray can. All other aerosols were sampled by the standard aerosol mass concentration monitor.

**Relative humidity = 21%. Low experimental sensitivity, although well within error limits, may indicate poor sticking of particles due to low relative humidity.